Entangling photons through nonlinear response of quantum wells to ultrashort pulses

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We show that many-body correlations among excitons originating from the Pauli exclusion principle in a quantum well embedded inside a microcavity provide a possibility to produce pairs of entangled photons by ultrashort laser pulses with a yield of $\sim 10^{-2}$. The quantum-field theoretical two-particle density matrix in second quantization is used to calculate entanglement for arbitrary emission angles. Largest response can be expected at symmetric emission angles for resonances with the heavy-heavy and light-light two-exciton states with remarkably nontrivial dependence of entanglement on the emission angles and on the ellipticity parameters of the excitation. We show that the angle dependence can be tailored by means of the microcavity. Interestingly, the emitted entangled 2-photon states are always in a triplet state.

Current entangled-photon sources are mainly based on the parametric down-conversion (PDC) inside nonlinear crystals[1, 2], such as BBO crystals[3, 4]. These sources suffer from two serious limitations. First, since the far off-resonant three-photon scattering contains two far off-resonant virtual states, the entangled-photon production yield is very low[5, 6], which limits the brightness of entangled-photon sources based on nonlinear crystals, leading to low signal-to-noise ratios and long measurement times[7]. Second, PDC produces entangled photons with the twice as long wavelength as the pump photons, which limits the operating wavelength[5]. Therefore, the problem of alternative sources of entangled photons is of the great importance. Quantum dot (QD) structures have already been used for this[8, 9, 10] making use of the relaxation of two excitons into one bound biexciton on a QD, although QD structures cannot achieve the brightness of quantum well (QW) structures.

The current limitation of QD structures is the low operating temperature of about 4 to 30 K, which is mainly due to the decoherence arising from exciton-phonon and hyperfine interactions. QW structures face the additional decoherence source due to Coulomb interactions, even at low temperatures. We therefore investigate the possibility to use the short time response for the production of entangled photons at time scales for which the biexciton binding energy E_{xx} cannot be resolved anymore, i.e. $T \ll \hbar/E_{xx}$, and therefore the Coulomb interactions is negligible. In order for this method to be effective, a microcavity is required to extract quickly the entangled photons. In addition, the microcavity can be used to tailor the angle dependence of the entanglement. We show that the Pauli exclusion principle, which is instantaneous, is sufficient to produce entangled photons from a QW structure with a high yield of 10^{-2}. This method of producing entangled photons is radically different from the well-known method based on the bound biexciton state [8, 9, 11, 12, 13, 14, 15].

The excitation of a QW in a cavity by the external field and emission of the photons due to the radiative recombination are driven by the interaction of the QW with the photonic modes of the cavity. They are found by quantizing the electromagnetic (EM) field in the whole space while taking into account the one-dimensional (1D) spatial modulation of the refractive index $n(z)$ with z axis coinciding with the crystal growth direction. The states of the EM field are specified by $\widehat{k} = (\mathbf{k}_{\|}, \omega, h)$. Here $\mathbf{k}_{\|}$ is the in-plane wavevector, ω is the frequency, $h = s, p$ denotes the polarization state. In units $\hbar = 1$, $c = 1$ we present the quantized field as (see e.g. Ref. 16)

$$\mathbf{A} = \frac{1}{(2\pi)^{3/2}} \sum_{\widehat{k}} \boldsymbol{\epsilon}_{\widehat{k}}(z) \frac{1}{\sqrt{2\omega_{\widehat{k}}}} u_{\widehat{k}}(z) e^{i\mathbf{k}_{\|}\cdot\boldsymbol{\rho}} a_{\widehat{k}}^{\dagger} + \text{h.c.}, \qquad (1)$$

where $\boldsymbol{\epsilon}_{\widehat{k}}$ are the unit polarization vectors ($\boldsymbol{\epsilon}_s$ lies in the plane of QW), $u_{\widehat{k}}(z)$ is the spatial distribution of the field along the z-axis found as the solution of the respective 1D scattering problem, $\boldsymbol{\rho}$ is the coordinate in (x, y) plane and $a_{\widehat{k}}^{\dagger}$ is the photon creation operator. The summation over $\widehat{k}$ implies the integration over the continuous quantum numbers and summation over the discrete ones.

Entanglement of the photons produced in the course of the radiative relaxation of the pumped semiconductor is found considering the two-photon density matrix

$$\rho_{\widehat{q}_1, \widehat{q}_2}^{\widehat{k}_1, \widehat{k}_2}(t) = \langle \Psi(t)| a_{\widehat{q}_1}^{\dagger} a_{\widehat{q}_2}^{\dagger} a_{\widehat{k}_1} a_{\widehat{k}_2} |\Psi(t)\rangle, \qquad (2)$$

where $|\Psi(t)\rangle$ is the state of the semiconductor-photon system. In lowest-order perturbation theory non-vanishing terms result from the contribution of the two-photon states into $|\Psi(t)\rangle$, leading to

$$\rho_{\widehat{q}_1, \widehat{q}_2}^{\widehat{k}_1, \widehat{k}_2}(t) = \Psi_{\widehat{q}_1, \widehat{q}_2}^*(t) \Psi_{\widehat{k}_1, \widehat{k}_2}(t) \qquad (3)$$

with $\Psi_{\widehat{k}_1, \widehat{k}_2}(t) = \langle 0 | a_{\widehat{k}_1} a_{\widehat{k}_2} | \Psi(t) \rangle$, where $|0\rangle$ is vacuum of the combined system. The free dynamics in the interaction picture is given by

$$\Psi_{\widehat{k}_1, \widehat{k}_2}(t) = \langle 0 | a_{\widehat{k}_1}(t) a_{\widehat{k}_2}(t) \mathcal{T}_+ \exp \left\{ -i \int_0^t dt' \widetilde{H}_{int}(t') \right\} |0\rangle, \tag{4}$$

where $a_{\widehat{k}}(t)$ and $\widetilde{H}_{int}(t)$ are the photon annihilation operator and the Hamiltonian of the light-matter interaction, respectively, and $\mathcal{T}_+$ is the time-ordering operator.

The low energy excitations are conveniently accounted by introducing the exciton operators according to $|\mu\rangle = B_\mu^\dagger |0\rangle$, where $|\mu\rangle$ is the hole-electron pair state either bound or unbound corresponding to energy E_μ, i.e. $H_{SC} |\mu\rangle = E_\mu |\mu\rangle$ with H_{SC} being the Hamiltonian of the nonperturbed semiconductor. For simplicity we assume that the QW can be approximated by a 2D plane situated at $z = z_0$. In this case the exciton states are characterized by the spin states of the hole and the electron constituting the pair, the center of mass momentum in the plane of the well, $\mathbf{K}$, and other quantum numbers, n_μ, so that $|\mu\rangle = |\sigma_\mu, s_\mu, \mathbf{K}_\mu, n_\mu\rangle$. Denoting by $\phi_\mu(\mathbf{x}, \mathbf{x}')$ the exciton (hole-electron) wave function corresponding to the state μ we represent the exciton operator as $B_\mu = \int d\mathbf{x} \, d\mathbf{x}' \, \phi_\mu^*(\mathbf{x}, \mathbf{x}') c_{s_\mu}(\mathbf{x}') v_{\sigma_\mu}(\mathbf{x})$, where c_s and v_σ are the annihilation operators for the electrons with the spin s and the holes with the spin σ, respectively. In terms of the exciton operators the light-matter interaction Hamiltonian is $H_{int} = \sum_\mu \left(\bar{\mathcal{A}}_\mu B_\mu + \mathcal{A}_\mu B_\mu^\dagger \right)$ with $\mathcal{A}_\mu$ and $\bar{\mathcal{A}}_\mu$ defined as convolutions of $\mathbf{A}(\mathbf{x})$ with $\mathbf{d}_{\sigma_\mu, s_\mu} \phi_\mu(\mathbf{x}, \mathbf{x})$ and its conjugate, respectively. Here $\mathbf{d}_{nn'} = e/m \langle n | \mathbf{p} | n' \rangle$ with $\langle n | \mathbf{p} | n' \rangle$ is the matrix element of the momentum operator between the bands n and n'.

In order to distinguish between the processes of the excitation of the semiconductor and the radiative exciton recombination we separate the contributions of the quantized field of emitted photons, $\mathbf{A}^{(q)}(\mathbf{x})$, and of the classical pumping field, $\mathbf{A}^{(cl)}(\mathbf{x})$, into the external field $\mathbf{A}(\mathbf{x}) = \mathbf{A}^{(q)}(\mathbf{x}) + \mathbf{A}^{(cl)}(\mathbf{x})$. In lowest-order perturbation theory we can neglect the processes of re-emission and reabsorption, thus obtaining

$$H_{int} \approx \sum_\mu \left(\bar{\mathcal{A}}_\mu^{(q)} B_\mu + \mathcal{A}_\mu^{(cl)} B_\mu^\dagger \right), \tag{5}$$

which we use in Eq. (4). Since in the low-intensity regime the higher-order terms of the light-matter interaction that affect the form of the photonic modes in the expression for the density matrix are negligible, the pump field is found by solving the respective initial value problem for the cavity alone without the QW. From now on we will omit the upper index for the quantized field.

Expanding the exponential term in Eq. (4) we obtain various terms depending on ordering of the operators B and $B^\dagger$. We leave only the terms describing the response along the directions different from the direction of the incident excitation field, where it is not blurred by the non-scattered field and by the linear (single-photon) response. Using the δ-functional approximation for the z dependence of the exciton wave function the two-photon amplitude of the outgoing photons can be presented as

$$\Psi_{\widehat{k}_1, \widehat{k}_2}(t) = u_{\widehat{k}_1}(z_0) u_{\widehat{k}_2}(z_0) \boldsymbol{\epsilon}_{\widehat{k}_1}(z_0) \cdot \overleftrightarrow{M}_{\mathbf{k}_1, \mathbf{k}_2}(t) \cdot \boldsymbol{\epsilon}_{\widehat{k}_2}(z_0), \tag{6}$$

where the tensor $\overleftrightarrow{M}_{\mathbf{k}_1, \mathbf{k}_2}(t)$ depends only on the direction of propagation of the outgoing photons but neither on their polarizations nor on the choice of the solutions of the scattering problems [see the supplement for the explicit form of $\overleftrightarrow{M}_{\mathbf{k}_1, \mathbf{k}_2}(t)$]. This information and the effect of the photonic density of states modified by the cavity are contained in the amplitudes $u_{\widehat{k}_{1,2}}(z_0)$. If the field distribution inside the cavity has the maximum near z_0 this results in accordingly amplified two-particle amplitudes $\Psi_{\widehat{k}_1, \widehat{k}_2}$. On the contrary, if for one polarization, s or p, the amplitude is significantly smaller comparing to the other this can be easily shown to imply significant decrease of the photon entanglement and so on.

The important property of the two-photon amplitudes follows from the conservation of the total in-plane momentum. Assuming the normal incidence of the excitation field this leads to the important restriction on the two-photon amplitudes $\Psi_{\widehat{k}_1, \widehat{k}_2} \propto \delta(\mathbf{k}_{1,\parallel} + \mathbf{k}_{2,\parallel})$. Since the QW is invariant only with respect to in-plane translations the restrictions is imposed only on the in-plane component of the wave vectors of the outgoing photons.

As Eq. (3) shows, the perturbation theory produces the density matrix corresponding to a pure state of the two photon system. Thus we can directly apply the standard machinery for evaluating entanglement of two photons as the von Neumann entropy of the reduced density matrix. The dependence of entanglement on the direction of propagation of the outgoing photons is very complex. In order to consider the main features of entanglement of the emitted photons, we discuss the details of the case of a single QW without the cavity, leaving the details of the

case including cavity to another paper. In the case without cavity one obtains a compact expression for the reduced single-photon density matrix

$$\rho_{\boldsymbol{\epsilon}}^{\boldsymbol{\epsilon}'}(t;\mathbf{k}_1,\mathbf{k}_2) = \boldsymbol{\epsilon} \cdot \overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)\big(\overleftrightarrow{1} - \widehat{\mathbf{e}}_2 \otimes \widehat{\mathbf{e}}_2\big)\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}^{\dagger}(t) \cdot \boldsymbol{\epsilon}', \tag{7}$$

where $\overleftrightarrow{1}$ is the unit tensor and the argument of $\rho_{\boldsymbol{\epsilon}}^{\boldsymbol{\epsilon}'}$ shows the dependence of the reduced density matrix on the wave vectors of the *pair* of the outgoing photons. The eigenvalues of the reduced density matrix determine entanglement as $E_N = -\widetilde{\rho}_1 \log_2(\widetilde{\rho}_1) - \widetilde{\rho}_2 \log_2(\widetilde{\rho}_2)$ with $\widetilde{\rho}_{1,2} = \rho_{1,2}/(\rho_1 + \rho_2)$. There are two terms in $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$, which describe different physical origins of entanglement of the emitted photons. One term describes the creation of entanglement due to the Pauli exclusion principle while another term accounts for the effect of the Coulomb interaction. Staying in the ultrashort time limit, we can neglect the effect of the Coulomb interaction.

For the resonance with heavy-hole excitons the non-normalized eigenvalues of the single-particle density matrix are proportional to the solutions of

$$\rho^2 - \frac{\rho}{2}\left\{\cos(2\chi)\sin^2(\beta)\sin^4(\theta) + \big[1 + \cos^2(\beta)\big]\right.$$
$$\left. \times \big[1 + \cos^2(\theta)\big]^2\right\} + \sin^4(\beta)\cos^4(\theta) = 0, \tag{8}$$

where β and χ are the polar and azimuthal angles on the Poincare sphere describing the polarization state of the excitation field, [17] $A_+ = e^{i\chi/2}\cos(\beta/2)$ and $A_- = e^{-i\chi/2}\sin(\beta/2)$. So that $\beta = 0, \pi, \pi/2$ correspond to the left and right circular and linear polarizations, respectively, and $\chi/2$ is the angle between the axis of the ellipse of polarization and the projection of $\mathbf{k}_1$ on the plane of the QW. Since both excitons have the same energy, the emission has a maximum at symmetric angles, i.e. when $\theta_1 = \theta_2$ (see Fig. 1a).

The dependence of entanglement on the direction of the outgoing photons and on the parameters of the excitation pulse is relatively simple in this case. Generally, the maximum entanglement is reached near $\theta = 0$ for the linear polarization where the non-normalized eigenvalues are $\rho_{1,2} = [1 \pm \cos(\beta)]^2$. For linearly polarized excitation field, $\beta = \pi/2$, the pairs are maximally entangled $E_N = 1$ while with decreasing the degree of ellipticity entanglement decreases. As the special feature the flat maximum near $\theta = 0$ should be emphasized. There $\partial E/\partial\theta \propto \sin^3(\theta)$ for arbitrary polarization of the excitation field. Moreover, for linearly polarized light when $\chi = \pi/2$ entanglement reaches the maximum, $E_N = 1$, and is independent of θ (see Fig. 1b). It should be noted, however, that as follows from Eq. (8) the eigenvalues of the reduced density matrix are $\rho_{1,2} \propto \cos^2(\theta)$ in this case, so the signal vanishes in the direction $\theta = \pi/2$.
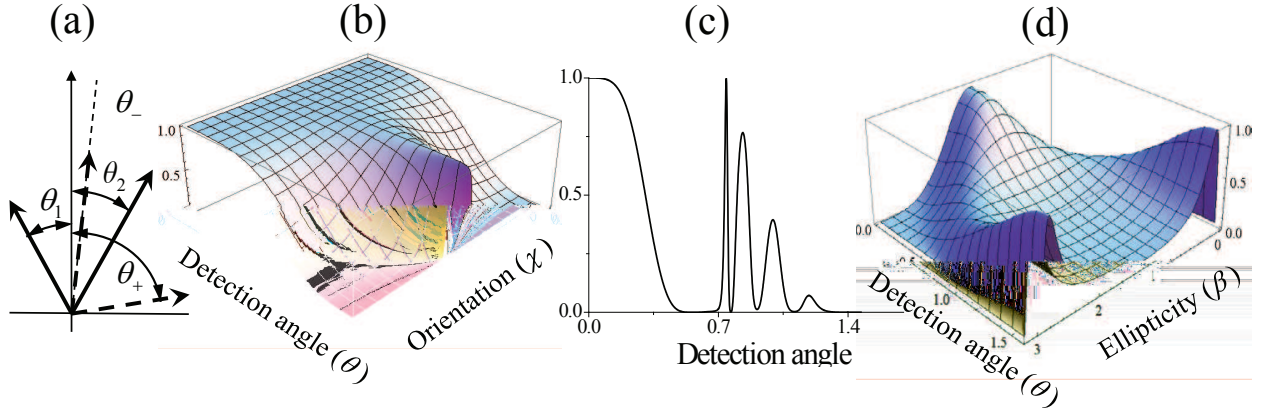

FIG. 1: (a) The emission directions. The symmetric case corresponds to the heavy-heavy and light-light two-exciton resonances. The asymmetric directions $\theta_2 = \theta_\pm$ correspond to the heavy-light resonance. (b) – (d) The Dependence of entanglement (vertical axes, scale from 0 to 1). (b) $E_N(\theta, \beta = \pi/2, \chi)$ in the vicinity of the heavy-hole exciton resonance. (c) $E_N(\theta, \beta = \pi/2, \chi = 0)$ in the presence of the cavity with 20 layers of the same optical width at $\theta = 0$, the index contrast is 1.5, the heavy-hole exciton resonance is tuned at the middle of the first stop-band at $\theta = 0$. (d) $E_N(\theta, \beta, \chi = 0)$ near the light-hole exciton resonance.

Using Eq. (6) in the polarization basis the states are written as $|\Psi\rangle = \sum_{\epsilon,\epsilon'} \Psi_{\epsilon,\epsilon'} |\epsilon,\epsilon'\rangle$, where $\Psi_{\epsilon,\epsilon'}$ are found convoluting the polarization vectors of the outgoing photons ϵ and ϵ' with $\overleftrightarrow{M}_{\hat{k}_1,\hat{k}_2}$. Along the direction $\theta \approx 0$, where $E_N = 1$ can be reached, we obtain

$$|\Psi\rangle \propto -e^{i\chi} [1 - \cos(\beta)] |+\rangle|+\rangle - e^{-i\chi} [1 + \cos(\beta)] |-\rangle|-\rangle, \tag{9}$$

where $|+\rangle$ ($|-\rangle$) is the state of a photon that is right (left) circularly polarized. As θ is increased, the entanglement is reduced down except for the case $\beta = \pi/2$, $\chi = \pi/2$, where the state is of the same structure as for $\theta = 0$, i.e. $|\Psi\rangle \propto |+\rangle|+\rangle-|-\rangle|-\rangle$, as long as $\theta < \pi/2$. Along $\theta = \pi/2$ the two-photon state is $|\Psi\rangle \propto [\cos(\chi)-i\cos(\beta)\sin(\chi)] |s\rangle|s\rangle$, i.e. the two-photon state is completely disentangled. Here the two-photon state is expressed in terms of the s and p polarization eigenstates. Varying the ellipticity of the incoming pulse away from linear polarization $\beta = \pi/2$, the entanglement is monotonously reduced down to $E_N = 0$ for $\beta = 0$. There the two-photon state reads $|\Psi\rangle = |\theta_1\rangle|\theta_1\rangle$ with $|\theta_1\rangle = (i\,|s\rangle + \cos(\theta)\,|p\rangle)/\sqrt{1 + \cos^2(\theta)}$, which is also completely disentangled. We would like to emphasize that all these states are triplet, that is transform according to the 3D representation of the rotation group.

Our calculations including the microcavity show that entanglement near $\theta = 0$ is not affected by the cavity. However, in general its angular dependence becomes highly non-trivial, approximately following that of transmittivities, as is illustrated in Fig. 1c. These properties are very useful for tailoring the angle dependence of the emission of the entangled photons. The details of the cavity model will be discussed elsewhere.

For the photons in resonance with the light-hole excitons $(\omega_1 = \omega_2 = E_l)$ the directional dependence of entanglement is more complex. The reason is the interaction of obliquely propagating p polarized photons with the light hole excitons with the zero projection of the total spin. The characteristic equation in this case differs from Eq. (8) by the term $X \{ X [\rho - 1 + \sin^2(\beta)\sin^2\chi] + 2\cos(\chi)\cos^2(\theta) [\rho - \sin^2(\beta)] \}$ in the right-hand side with $X = 8\sin(\beta)\sin^2(\theta)$. This term becomes important for oblique directions thus yielding a richer structure of the angular dependence of entanglement, as shown in Fig. 1d.

For the photons in resonance with heavy- and light-hole excitons, corresponding to $\theta_1 \neq \theta_2$, there is only one resonance, when the two-exciton state is made of light-hole and heavy-hole excitons. Respectively, there are only two resonant directions with $\theta_1 \neq \theta_2$ with

$$\sin(\theta_\pm) = \sin(\theta_1) \left(\frac{E_l}{\Delta_{hl}} \right)^{\pm 1}. \tag{10}$$

The reduced single-photon density matrix is determined by Eq. (7) with $\overleftrightarrow{M}_{\hat{k}_1,\hat{k}_2} \propto A_+ A_- \left(\overleftrightarrow{1} - \hat{e}_z \otimes \hat{e}_z \right)$. Its non-normalized eigenvalues are 1 and $\cos^2(\theta)$. As a result, entanglement monotonously decreases from 1 to 0 as θ changes from 0 to $\pi/2$ while the direction of the detection of the second photon is determined by Eq. (10). For sufficiently large detection angles θ such that $\sin(\theta) > \Delta_{hl}/E_l$ only one resonant direction corresponding to θ_- remains. Interestingly, in the asymmetric case entanglement is independent of the the polarization of the excitation field, however, $\overleftrightarrow{M}_{\hat{k}_1,\hat{k}_2}$ vanishes if the excitation pulse is circularly polarized.

Studying entanglement would not be complete without considering yield, which is defined as the ratio of the energy flux carried by the entangled pairs of the photons to the flux of the excitation field. For practical purposes it is more convenient to use an alternative definition $Y = N_{out}/N_{in}$, where N_{out} and $N_{in} \sim (\Phi/\hbar\Omega)^2$ are the number of outgoing and incoming *pairs*, respectively, with Φ being the total flux of the external field and Ω being its frequency in the stationary frame. Calculating the trace of the single-particle density matrix over the polarization quantum numbers we find (in SI units)

$$Y \sim \frac{4\pi^9 E_x}{3\gamma^3 S} (\rho_1 + \rho_2) \left(\frac{T|Q|^2}{\hbar\Omega c^3 \epsilon_0^2} \right)^2, \tag{11}$$

where E_x is the exciton energy, $\rho_{1,2}$ are the non-normalized eigenvalues of the density matrix defined by the equations studied above, S is the area of the excitation spot, T is the duration of the excitation pulse, Q is the common interband dipole moment, and ϵ_0 is vacuum permittivity. The inverse dependence on the pump area is clear since the dense excitation more pronounced is the effect of the exclusion principle. The dependence on the pulse duration is the consequence of the semiconductor response determined by the polarizations of the linear response rather than by the energy input. Substituting the values typical for GaAs, $\gamma = 1.5$ meV, $\hbar\Omega \approx E_x = 1.5$ eV, $Q = 1.3 \cdot 10^{-24}$ kg $\cdot$ m/s (see e.g. Ref. 18) and using $T = 100$ fs, $S = \pi(20)^2$ μm^2 we find $Y \sim 0.02$. Such high value of yield is the result of the resonant transitions between the many-particle states.

In conclusion, we have studied the basic mechanism of emission entangled photons by a semiconductor quantum well excited by a short pulse. We have developed a "kinematic" theory accounting the effect of the exclusion principle.

The dependence of entanglement on the detection angle and on the polarization of the external field is shown to be highly nontrivial. We have estimated yield of the considered process and have found it to be rather high owing to the resonant transitions between different states.

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SUPPLEMENTAL INFORMATION

In the lowest non-vanishing order of the perturbation theory Since the tensor $\overleftrightarrow{M}_{\widehat{k}_1,\widehat{k}_2}(t)$ does not depend on the polarization states of the outgoing photons we omit the hats over k's and write it solely in terms of the wave vectors of the outgoing photons

$$\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t) = \frac{2\pi}{4\sqrt{\omega_{k_1}\omega_{k_2}}}\widetilde{\phi}_{\mu_1}(0)\widetilde{\phi}_{\mu_2}(0)\int\limits_0^t dt_1 e^{-i(\omega_{k_1}-\omega_{k_2})(t-t_1)}\int\limits_0^{t_1} dt_2\, e^{-iE_{\mu_1}(t_1-t_2)}$$

$$\Big[e^{-i\omega_{k_2}(t_1-t_2)}\delta(\mathbf{K}_{\mu_1}-\mathbf{k}_{1,\parallel})\delta(\mathbf{K}_{\mu_2}-\mathbf{k}_{2,\parallel})\,\mathbf{d}_{\sigma_{\mu_1},s_{\mu_1}}\otimes\mathbf{d}_{\sigma_{\mu_2},s_{\mu_2}}$$

$$+e^{-i\omega_{k_1}(t_1-t_2)}\delta(\mathbf{K}_{\mu_2}-\mathbf{k}_{1,\parallel})\delta(\mathbf{K}_{\mu_1}-\mathbf{k}_{2,\parallel})\,\mathbf{d}_{\sigma_{\mu_2},s_{\mu_2}}\otimes\mathbf{d}_{\sigma_{\mu_1},s_{\mu_1}}\Big]\mathcal{G}_{\mu_1,\mu_2}(t_2), \tag{12}$$

where $\otimes$ denotes the tensor product so that in a particular coordinate system $(\mathbf{d}\otimes\mathbf{d}')_{ij} = d_i d'_j$, $\omega_k = k$ is the photon energy and E_μ are the energies of the hole-electron pair states. The effect of the semiconductor nonlinear response is described by the function $\mathcal{G}_{\mu_1,\mu_2}(t)$, which we present as a sum of the instantaneous term and the memory term

$$\mathcal{G}_{\mu_1,\mu_2}(t) = \left\langle B_{\mu_1}B_{\mu_2}B_{\nu_3}^\dagger B_{\nu_4}^\dagger\right\rangle P_{\nu_3}^{(1)}(t)P_{\nu_4}^{(1)}(t) + i\int\limits_0^t dt' \left\langle B_{\mu_1}B_{\mu_2}e^{-iH_{SC}(t-t')}D_{\nu_3,\nu_4}^\dagger\right\rangle P_{\nu_3}^{(1)}(t')P_{\nu_4}^{(1)}(t'), \tag{13}$$

where[19] $D_{\nu_3,\nu_4}^\dagger = [B_{\nu_3}^\dagger,[B_{\nu_4}^\dagger,H]]$ and $P_\nu^{(1)}(t)$ are the exciton polarizations of the linear response created by the action of the external (classical) field

$$P_\nu^{(1)}(t) = -i\int\limits_0^t dt'\, e^{-iE_\nu(t-t')}\mathcal{A}_\nu^{(cl)}(t'). \tag{14}$$

Neglecting the memory term (or the contribution of the Coulomb interaction) the tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ is directly expressed in terms of A_+ and A_-, the amplitudes of the left circular and right circular components of the excitation pulse, respectively,

$$
\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t) = \frac{(2\pi)^5}{4\sqrt{\omega_1\omega_2}}\,\delta(\mathbf{k}_{1,\parallel}+\mathbf{k}_{2,\parallel})\left(A_-^2\,\widehat{\mathbf{e}}_+\otimes\widehat{\mathbf{e}}_+ + A_+^2\,\widehat{\mathbf{e}}_-\otimes\widehat{\mathbf{e}}_-\right)\left[W_{hh}(t)+\frac{1}{9}W_{ll}(t)\right]
$$
$$
+\frac{2}{3}\left(\widehat{\mathbf{e}}_+\otimes\widehat{\mathbf{e}}_- + \widehat{\mathbf{e}}_-\otimes\widehat{\mathbf{e}}_+\right)A_+A_-\left[W_{hl}(t)+W_{lh}(t)\right] \tag{15}
$$
$$
-\frac{8}{9}\widehat{\mathbf{e}}_z\otimes\widehat{\mathbf{e}}_z A_+A_- W_{ll}(t).
$$

Here the indices h and l denote $|\sigma| = 3/2$ and $|\sigma| = 1/2$, respectively. The time dependence is described by $W_{\sigma_1,\sigma_2}(t) = \mathcal{I}_{\sigma_1,\sigma_2}\left[w_{\sigma_1,\sigma_2}(\omega_1,\omega_2;t)+w_{\sigma_1,\sigma_2}(\omega_2,\omega_1;t)\right]$ with

$$
w_{\sigma_1,\sigma_2}(\omega_1,\omega_2;t) = \int_0^t dt_1 e^{-i(\omega_1+\omega_2)(t-t_1)}\int_0^{t_1} dt_2 e^{-i(\omega_1+E_x)(t_1-t_2)}e^{-iE_{xx}t_2}, \tag{16}
$$

where $E_x = E_{\sigma_1}$ and $E_{xx} = E_{\sigma_1} + E_{\sigma_2}$ denote single-exciton and two-exciton energies, respectively. The intensity of the exciton-light interaction is quantified by $\mathcal{I}_{\sigma_1,\sigma_2} = |Q|^4\left|\widetilde{\phi}_{\sigma_1}(0)\widetilde{\phi}_{\sigma_2}(0)\right|^2\int d\mathbf{q}\,|\widetilde{\varphi}_{\sigma_1}(\mathbf{q})|^2\,|\widetilde{\varphi}_{\sigma_2}(\mathbf{q})|^2$ with $\widetilde{\varphi}_\sigma(\mathbf{q})$ being the Fourier transform of the exciton wave function. Here and in Eq. (15) we have taken into account the structure of the valence band in the semiconductors with the point symmetry T_d and have introduced the common interband dipole moment $Q = -ie/m\,\langle X|\,p_x\,|\Gamma_1\rangle$ (see e.g. Ref. 18).

The important result immediately following from Eq. (15) is that if the external excitation field is circularly polarized then the emitted photons are disentangled. Indeed, in this case the tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ is represented as a tensor product and, hence, so is the single-particle density matrix, i.e. it corresponds to a pure state.

Representation (15) explicitly shows the tensor $\overleftrightarrow{M}_{\mathbf{k}_1,\mathbf{k}_2}(t)$ as a superposition of the amplitudes of the radiative decay of different two-exciton states through two channels into the two-photon states. Due to the presence of different characteristic frequencies the time dependence of the total amplitude has a complex form, especially at the transitional regime with the duration proportional to inversed heavy-hole light-hole splitting. However, there are several resonances whose amplitudes increases with time and which define the long time response and respectively the time dependence of entanglement. In order to extract these resonances we consider the long time limit in the spirit of the Wigner-Weisskopf approximation [20] extending the limits of integrations over time to infinity. This yields

$$
w(t) \propto e^{-iE_{xx}t}\delta\left[E_{xx}(1+\alpha-b)-E_x(1+\alpha)\right]\delta\left[\omega_1+\omega_2-E_{xx}\right]. \tag{17}
$$

Here we have taken into account the momentum selection rule and have introduced $\alpha = \sin(\theta_1)/\sin(\theta_2)$ and b stands for either 1 or α for $w(\omega_1,\omega_2)$ and $w(\omega_2,\omega_1)$, respectively, that is depending on particular exciton-photon channel. As follows from Eq. (17) only such terms in Eq. (15) contribute into the long limit which satisfy the special resonant condition. The total energy of the emitted pair must be equal to the energy of the two-exciton state. Additionally there is the special "kinematic" requirement imposed on the energies of the involved single- and two-exciton states.